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A METHOD FOR IMPROVING ACCURACY OF AIR PARTICULATE ACTIVITY MEASUREMENTS

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Bureau of Medicine and Surgery, Navy Department
Research Project MR005.14-3002-4.07

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SUMMARY PAGE

THE PROBLEM

To investigate the nature of particulate radioactivity of the air in a nuclear-powered submarine, and to estimate whether there might be any possible long-term effects on personnel caused by radiation associated with the air particles.

FINDINGS

A new method was devised for obtaining more accurate estimates of the instantaneous air particulate activity, which greatly increases the precision of these measurements. Results indicate that under normal operating conditions this activity is well within perscribed conservative limits.

APPLICATION

The information presented in this report will be of value to medical and other personnel charged with protecting the health of military personnel. In a practical way, the new measuring method permits more flexibility in the processing of air samples and also makes possible better counting statistics.

ADMINISTRATIVE INFORMATION

This investigation is reported under Bureau of Medicine and Surgery Research Task MR005.14-3002-4, Field Evaluation of Products and Equipment Affecting Submarine Habitability. The present report is No. 7 on this Subtask and was submitted by the author in partial completion of the requirements for qualification as a Submarine Medical Officer.

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ABSTRACT

In order to investigate the air particulate radioactivity in nuclear-powered submarines more precisely than has been done heretofore, four members of the Blue Crew of a nuclear-powered submarine, the USS ROBERT E. LEE SSB(N)-601, were subjected to whole-body gamma radiation tests before they had any known contact with radioactivity or nuclear reactors. They were similarly tested after participating in a 60-day patrol.

Preliminary analysis of the two sets of data indicates the absence of any internally-deposited radionuclides. It thus appears that air particulate activity at its present levels and under normal operating conditions is well within perscribed conservative limits against external and internal beta radiation. More complete conclusions will be possible after computer analysis of the data.

A METHOD FOR IMPROVING ACCURACY OF AIR PARTICULATE ACTIVITY MEASUREMENTS*

INTRODUCTION

The medical officer attached to Submarine Squadron TEN became interested in investigating the air particulate activity in the enclosed atmosphere of a nuclear-powered submarine, and in determining whether there might be minute quantities of long-lived gamma radiation being deposited in the bodies of the personnel serving in this type of ship. He was aware that a different method of measuring the air particulate activitv would be necessary since the equipment currently aboard these submarines will only count very high intensity gamma activity.

The present method used aboard nuclear submarines for the determination of the particulate air activity of samples collected with the (HD-251 (SN-3)UD) or "Cadillac" apparatus, assumes that the particulate air activity has a rather long half life. By using this assumption, particulate air activity is estimated by dividing the value of the activity counted on the collecting filter media by the total volume of air passed through the filter media during the sampling period. Studies of decay of activity collected on filters** indicate that the air particulate activity has a relatively short half-life of about 35 minutes. A large fraction of this activity is beta radiation but on large-area filter samples collected over periods of several hours, alpha radiation in the range of 45 to 50 $\mu\mu c/ft^2$ can also be detected. Activity with this half-life, containing appreciable amounts of alpha radiation, is compatable

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* The opinions or assertions contained herein are the private ones of the author and are not to be construed as official or reflecting the views of the Navy Department or the naval service at large.

** These figures are based on studies of the air particulate activity on board the USS ROBERT E. LEE (SSB(N)-601) during the prolonged submergence period of the ship's third patrol, November-December, 1961.

with the activity produced by radon, thoron, and their daughter products (1, 3). These isotopes result from the decay of naturally occurring radium and thorium found in minute quantities in the materials of construction of the ship and larger quantities of radium found on the luminous dials of watches and gages which were brought on board undetected. Once a submarine has been isolated from the earth's atmosphere for several days, the rate of production and the rate of decay of the various isotopes become about equal and the average air particulate activity becomes about constant.

Since the half-life of the activity is in fact relatively short, and since no correction is made for decay of activity on the filter during the actual sampling period or during the delay period between the end of the sampling and the beginning of counting, the present method of calculation will yield consistently low estimates of the air particle activity. Moreover, the errors which result may vary from day to day unless the time of sampling and the delay time after sampling are both held constant and a constant correction factor is applied. In practice, times may vary from measurement to measurement and operator to operator. Also, standard counting techniques (2, 3) do not specify fixed sampling and delay times before counting and suggest no correction factors for errors resulting from variations in these times. The advantage of improved counting statistics which might result from rather long sampling times is also lost unless a correction factor is applied because the error caused by radioactive decay during sample collection becomes increasingly larger as sampling time rises.

THEORETICAL CONSIDERATIONS

In order to obtain more precise results during daily measurements and to anticipate trends in the air particulate activity with more accuracy, a mathematical correction may be applied to the value of activity which is obtained by the usual method of estimation. The following assumptions are made:

(a) The volume of air passed through the collecting filter per unit of time is essentially constant. This assumption was checked by plotting a curve of air volume through the filter versus time. Over a period of 90 minutes, an essentially straight line was obtained with the HD-251 air sampling apparatus, even though the filter was darkened by a fairly heavy collection of particles. This would indicate that the resistance to air flow through the filter element results primarily from the construction of the filter element itself and is not greatly influenced by the trapping of the air particles in the filter interstices with subsequent reduction of the flow area.

(b) The order of magnitude of the air particle activity in μc/ml is assumed to vary little from day to day. Since the half-life of the particulate activity is about 35 minutes, a rather steady source of air contamination in the form of natural radon and thoron production in the materials of construction of the ship, as well as minute primary and associated system piping leaks, is necessary. If this were not so, the air particulate activity would decay away to undetectable levels within a few hours. The order of magnitude of the activity does in fact remain nearly constant during periods of prolonged submergence if reactor power level does not vary greatly.

To illustrate the nature of the errors introduced by neglecting the decay of the activity of the material which is collected, reference is made to Figure 1, a plot of activity detected on the collecting filter, versus time. For no decay of the collected particulate activity, a constant rate of air flow through the filter, and an essentially constant air activity during the period of collection, the total activity collected on the filter will vary linearly with respect to time, as represented by OA', and at time t_1, the total collected activity on the filter would correspond to a_0. However, since the activity accumulated on the filter is decaying at a rate determined by its half life and the quantity of active material present at any time, and since activity is collected from the air at a constant rate, it becomes evident that the rate of change of activity with respect to time will gradually decrease to zero if sufficient time for collection is allowed. When this situation prevails, the

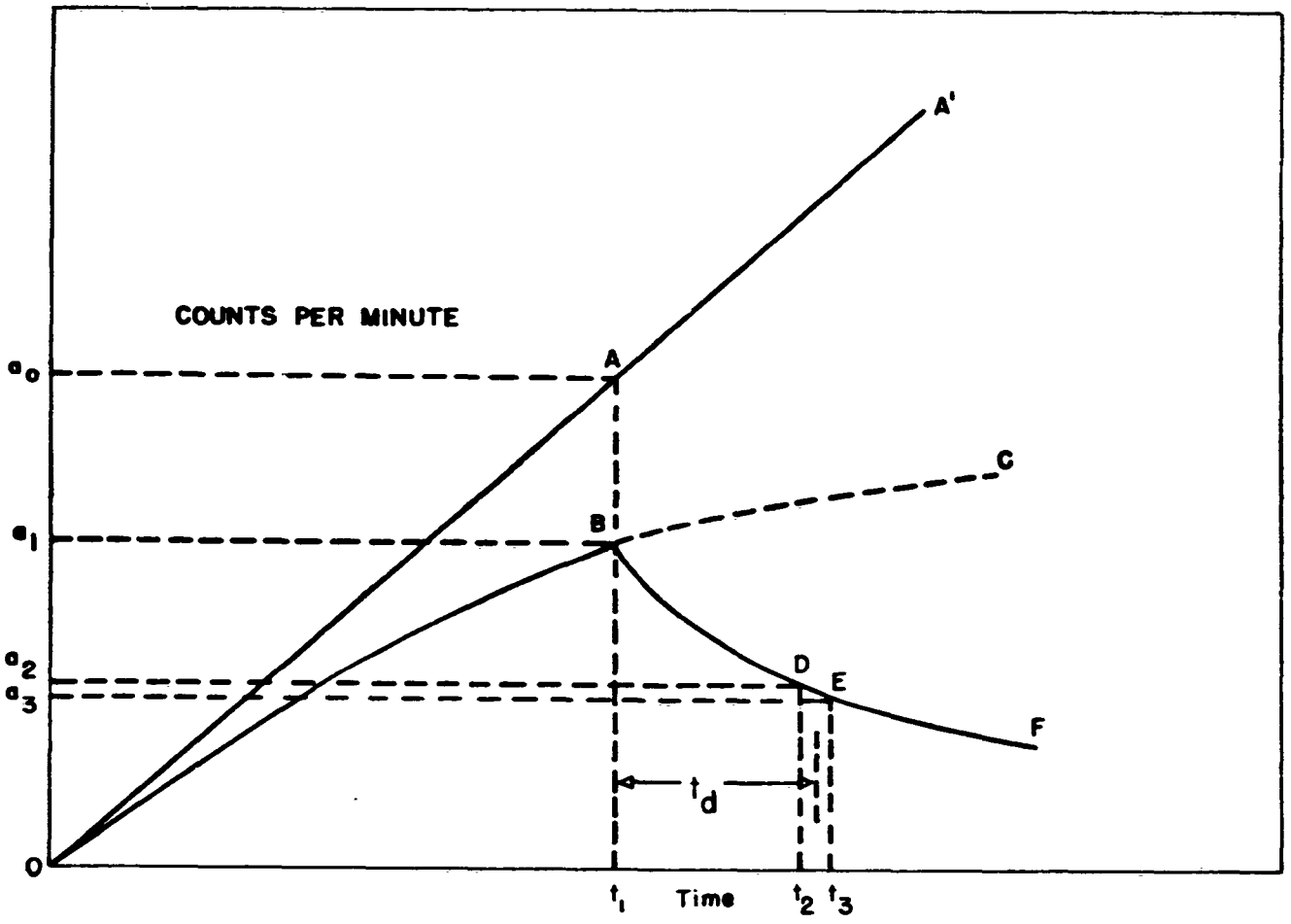

Figure I. Activity of Particulate Air Filter Samples versus Time

rate of addition of new activity by the passage of air through the filter will just equal the rate at which activity disappears from the filter by the process of radioactive decay. This sequence is shown by OBC, where the time rate of change of activity is approaching zero at point C.

If the sample collection is completed at time t_1, the corresponding activity on the filter will be a_1', a value somewhat lower than a_o, the activity which would be present if no decay had taken place. Moreover, since air flow through the filter has ceased, no new activity is being added to the sample. The particulate activity on the filter, however, continues to decay at a rate determined by its half-life, as represented by BDEF.

If the filter is now placed in the counter-scaler pig and counted during the period of time between t_2 and t_3, the estimated value of the filter activity will be the mean between a_2 and a_3. When this mean value of the filter activity is divided by the total air volume passed through the filter between time zero and time t_1, the estimated air particulate activity will be considerably lower than the actual activity estimated by dividing activity a_o by the total air volume which passed through the filter up until time t_1. Also, the magnitude of the error involved will vary between operator and operator and from day to day as the sample collection time and the delay time before sample counting are varied.

METHOD OF CALCULATION

In an effort to obtain more accurate estimates of the instantaneous air particulate activity and increase the precision of the measurements, the following method of obtaining correction factors was devised:

Let **a** = the activity on the sample filter at any given time, in μc,
Let A_a = the instantaneous particulate activity of the air, μc/ml,

T_a = the half life of the particulate activity, min

V_t = the volume of air passing through the collecting filter per unit time, ml/min,

t = the time during which the sample collection occurs, min,

λ = the radioactive decay constant = $0.693/T_a$ min^{-1}.

Therefore, the rate of collection of activity on the filter = $A_a V_t$ μc/min, and the rate of decay of the activity on the filter = λa, μc/min, and

$$da/dt = A_a V_t - \lambda a.$$

Let $A_a V_t$ = a constant = b.

Thus $da/dt = b - \lambda a$,

and $da/(b - \lambda a) = dt$.

Since a = 0 when t = 0, and $a = a_1$ when $t = t_1$, the above equation may be integrated between these limits:

$$\int_0^{a_1} da/(b - a) = \int_0^{t_1} dt$$

yielding

$$(-1/\lambda)\ln (b - \lambda a)\Big]_0^{a_1} = t_1$$

or

$$b/(b - \lambda a_1) = e^{\lambda t_1},$$

where solving for a_1 and substituting the values for b and λ yields,

$$a_1 = (A_a V_t T_a/0.693)\ (1 - e^{-0.693 t_1/T_a})\ \mu c.$$

If no decay had occurred, the activity on the filter would have been a_o:

$$a_o = A_a V_t t_1\ \mu c.$$

Thus, to correct a measured value of a_1 to the value of a_o, which represents the total activity which has been collected, ignoring decay, during the time period between zero and t_1, the value of a_1 must be multiplied by a factor equal to a_o/a_1. Thus the correction factor, C_1, to correct for decay while collecting the sample is:

$$C_1 = a_o/a_1 = (A_a V_t t_1/A_a V_t T_a)\frac{0.693}{(1 - e^{-0.693 t_1/T_a})}$$

or

$$C_1 = (0.693 t_1/T_a)\quad (1 - e^{-0.693 t_1/T_a}).$$

Thus, for any given value of the half-life of the air particulate activity and time duration of sample collection, the value of C_1 may be calculated. The values of C_1 for several values of half-life and time duration of sample collection are listed in Table I:

TABLE I

Half Life of Particulate Air Activity, Min. (T_a) / Sample Collection Time, Min. (t_1)	20	30	40	60	100
20	1.39	1.25	1.19	1.12	1.07
40	1.85	1.53	1.39	1.25	1.14
60	2.38	1.85	1.61	1.39	1.22
80	2.96	2.20	1.85	1.53	1.30
100	3.58	2.56	2.10	1.68	1.39

For ease of estimation of the value of C_1, the values listed in the table above were plotted to yield the curves shown in Figure 2.

In order to correct for the decay of activity on the filter during the period of time between the completion of sample collection and the mean time of counting the sample, a second correction factor, C_2, may be derived. Reference to Figure 1 shows that the mean delay time after the completion of sampling may be expressed as t_d, where a fairly accurate extimate of t_d may be derived from the relation:

$$t_d = (t_2 + t_3)/2 - t_1.$$

Since, during the delay time, the activity on the filter decays exponentially, by reference to Figure 1, the mean values between the activities a_2 and a_3, $\overline{a_2 a_3}$, may be expressed as:

$$\overline{a_2 a_3} = a_1 e^{-0.693 t_d / T_a}$$

where λ, the decay constant has been expressed as $0.693/T_a$.

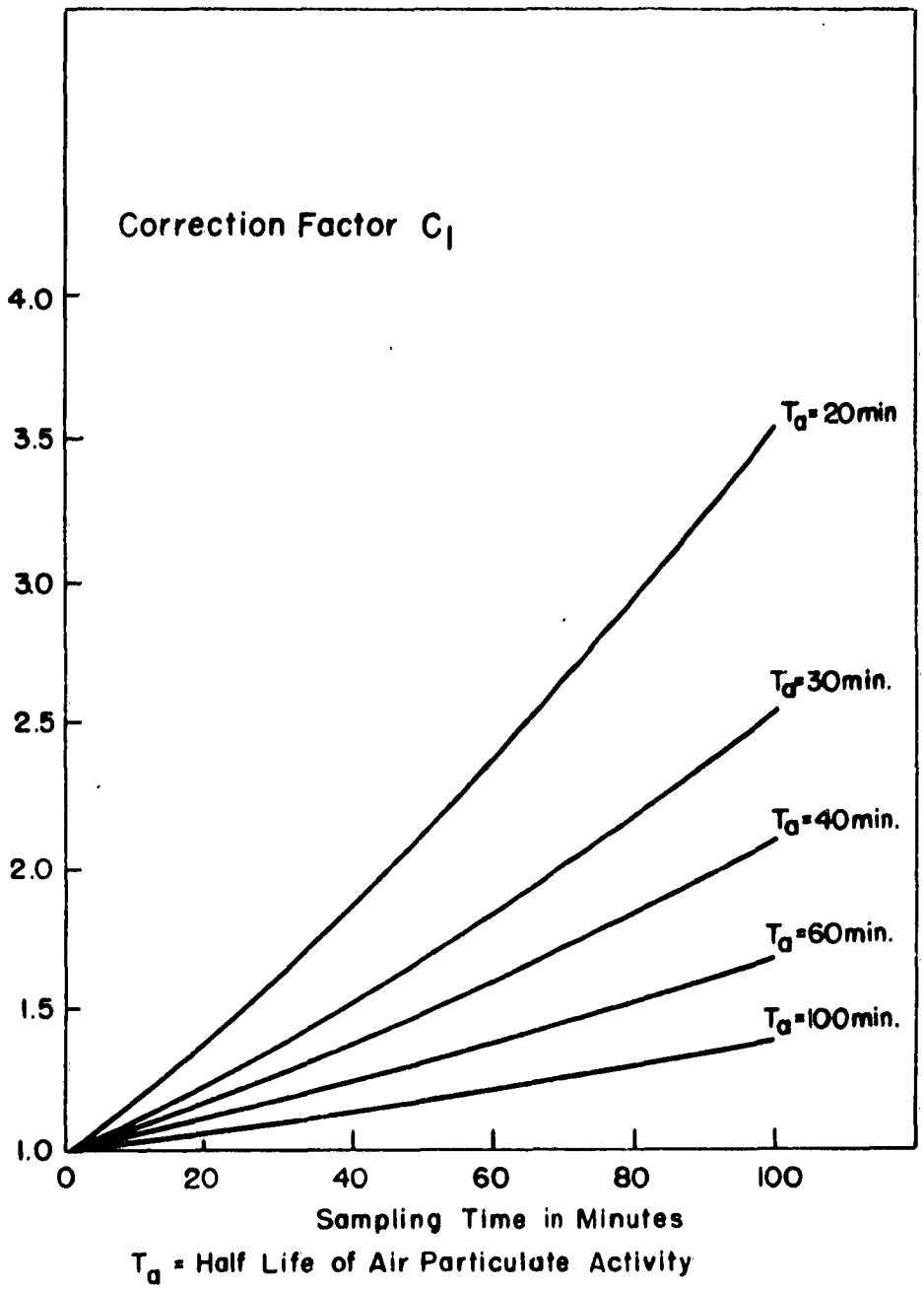

Figure 2. Correction Factor for Decay During Sampling
Air Particle Sampler HD-251

The correction factor for decay during the counting delay time may be expressed as C_2 where:

$$C_2 = a_1/\overline{a_2 a_3} = a_1/a_1 e^{-0.693 t_d/T_a} = e^{0.693 t_d/T_a}.$$

Thus, if the value of the activity actually observed on the filter is multiplied by C_2, a_1 becomes known; multiplying a_1 by C_1 will give the value of a_o, the total activity collected on the sample during the period of time between zero and t_1. A more correct value of A_a, the instantaneous air particle activity, may now be estimated from a_o and the known value of $V_t t_1$

The values of C_2 for several values of air particulate half-life and delay time after completion of sampling have been calculated and are listed in Table II.

TABLE II

Half Life of Particulate Air Activity, Min. (T_a)	5	10	15	20	30	45	70	100
Counting Delay Time, Min. (t_d)								
5	2.00	1.41	1.26	1.19	1.12	1.08	1.05	1.03
10	4.00	2.00	1.59	1.41	1.26	1.17	1.10	1.07
15	8.00	2.83	2.00	1.68	1.41	1.26	1.16	1.11
20	16.00	4.00	2.52	2.00	1.59	1.36	1.22	1.14
25	32.00	5.68	3.17	2.38	1.79	1.47	1.28	1.18
30	64.00	8.00	4.00	2.83	2.00	1.59	1.35	1.23

To facilitate estimation of C_2, the values calculated and listed in Table II have been plotted to yield the correction curves shown in Figure 3.

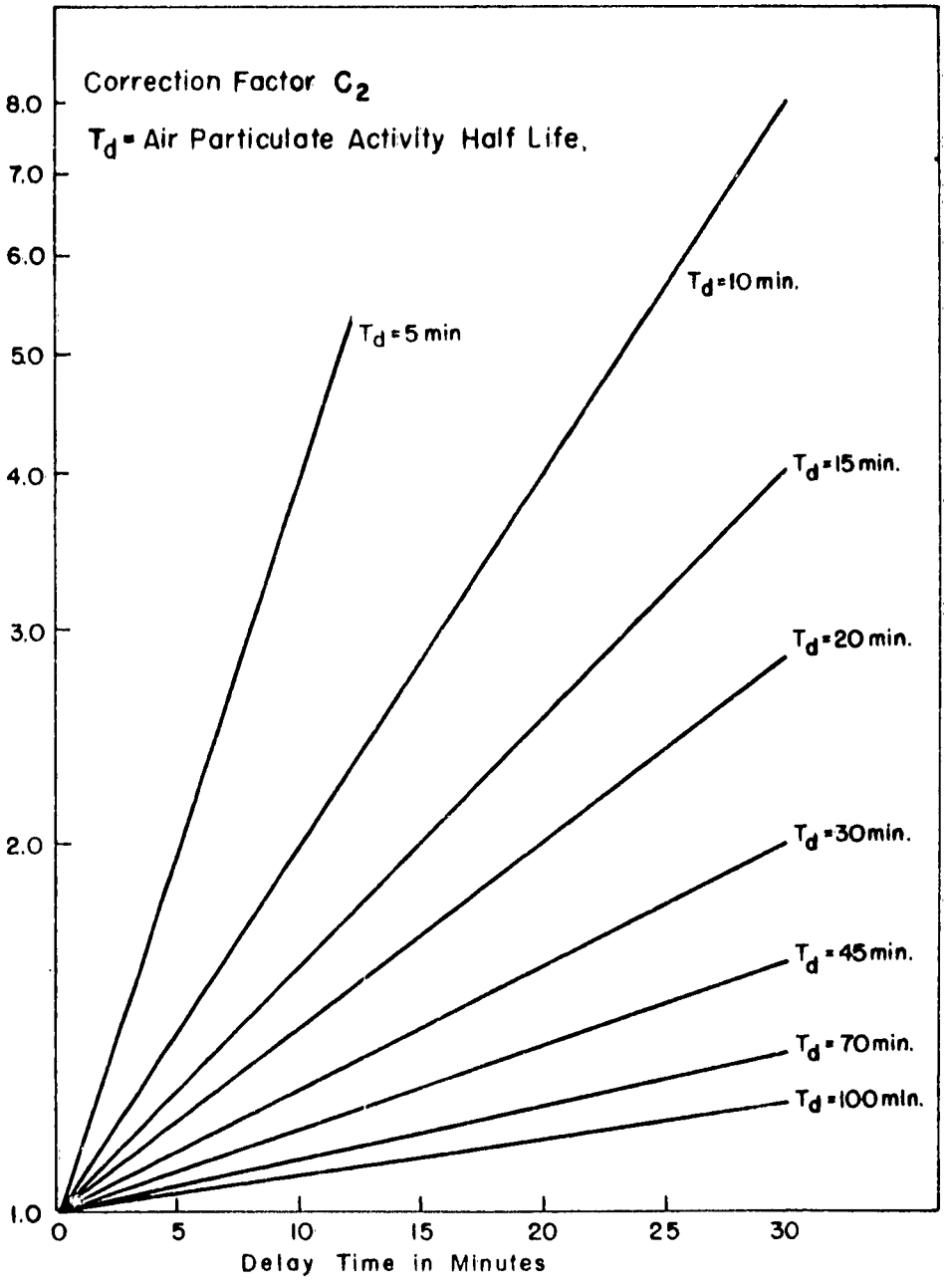

Figure 3. Correction Factor for Delay Time
Air Particle Sampler HD - 251

In order to estimate the corrected air particulate activity A_a, in μc/ml, the measurement of air activity is made in the usual manner and multiplied by the correction factors C_1 and C_2. Also, the following relation may be used:

Air Particulate Activity, μc/ml =

$$(C_s - C_b) \times 1/E \times 1/F \times 1/V \times C_1 \times C_2$$

where C_s = raw sample count, counts per minute

C_b = background count, counts per minute

E = scaler efficiency, counts per disintegration

F = conversion factor, 2.22×10^6 disintegration per minute per microcurie,

V = total volume of air passed through the filter during the collecting period, ml,

C_1 = correction factor for radioactive decay during the sampling period,

C_2 = correction factor for radioactive decay during the delay time before counting the sample.

By use of the appropriate times for the evaluation of C_1 and C_2, the sample collecting time and the delay time before counting can be varied without affecting the resultant estimated value of the air particulate activity. Also, a more meaningful comparison between activities obtained on different ships may be made. Better precision of values obtained with different sampling and delay times at the same activity level is also possible.

An added advantage can be obtained when these correction factors are applied. Better counting statistics are possible with samples of higher absolute activity, that is, with samples collected over relatively long sampling periods. It will be noted on Figure 1, that without decay corrections, the longer the period of sampling, the greater will be the error in the estimated value of the air particulate activity. In fact, with longer and longer collection times, lower and lower values of estimated particulate activity will be obtained since the calculation involves the division of uncorrected sample activity figures by increasingly larger air volume figures.

Although these corrections appear to be somewhat complex, once the half-life of the air activity is known, the use of Figures 2 and 3 will yield correction factors with little additional effort. The balance of the calculation is identical to that presently in use. When the approximate value of the sample count rate and the average background count rate are known, sample and background counting times may be estimated from available charts (2) in order to minimize statistical counting errors. Relatively long sample collecting times will also facilitate higher sample count rates and yield smaller statistical errors.

EFFECTS ON PERSONNEL

Air particulate radioactivity rarely if ever, rises above the conservative limits (1) set for U. S. Naval nuclear-powered ships. As mentioned previously, the isotopic nature of the activity routinely detected appears to be primarily radioactive daughter products of the decay of naturally occurring radium and thorium. These isotopes in most cases decay by beta particle emission and the counter-scaler detection of radioactivity on air particle filter specimens results mainly from beta activity. The efficiency of the counter scalers for the detection of gamma radiation, however, is only about one per cent of that for beta particle detection. Even though the apparent half-life of the activity is relatively short, it is possible that minute amounts of long-lived gamma emitting isotopes are present which escape detection with the present apparatus and methods. Since the air particulate activity is found throughout the entire enclosed submarine atmosphere, and the particles must settle on food and become ingested and inhaled to some extent, the question arises as to whether prolonged exposure of personnel to the atmosphere does not result in increasing body retention of isotopes which emit penetrating gamma radiation. If this situation could in fact be demonstrated, and the isotopic nature of the elements determined, much useful information might be obtained about the source of the isotopes and modes of attack to reduce the amounts present.

At present, no extremely sensitive gamma-radiation equipment is available on board submarines to measure the gamma ray spectrum emitted by the human body. Facilities have been developed (4) at the Brookhaven National Laboratory which make possible the detection of minute amounts of gamma emitting isotopes within the body. The measurement of the gamma radiation energy spectrum is also possible with this apparatus and thus particular isotopes may be identified by their characteristic gamma radiation spectrum. Briefly, whole-body gamma counting is carried out at Brookhaven in a thick-walled lead-, copper-, cadmium-, and steel-lined chamber which serves as shielding from outside sources of radiation. The subject to be counted sits within the chamber in close proximity to a 4" by 8" NaI (Tl) crystal detector mounted with three 3" photomultiplier tubes. Gamma radiation striking the NaI crystal causes minute scintillations which are picked up by the photomultiplier tubes, amplified, and fed to a 100-channel pulse-height analyzer. Analyzer circuits detect the various gamma energies present and indicate energy peaks which are characteristic of a particular gamma-emitting isotope. Counting statistics are relatively high since subjects are subjected to periods of counting as high as 30 minutes.

With the cooperation of the Medical Physics Division at Brookhaven, four members of the Blue Crew of the Polaris Submarine USS ROBERT E. LEE (SSB(N)-601) were subjected to whole-body counting before they had any known contact with radioactivity or nuclear reactors, other than natural background radiation and fallout from nuclear weapons tests. Just subsequent to a patrol of about 60 days duration, they were recounted in the same chamber at Brookhaven in an effort to detect any increase in long-lived gamma activity. Preliminary analysis of the two sets of data (5) indicates the absence of any internally-deposited radionuclides other than those normally occurring. The totals of gamma activity in all individuals were somewhat higher than the baseline readings but this increase was accounted for by the general rise in the world-wide levels of Cs^{137}. More complete data will be available after computer analysis.

It thus appears that air particulate activity at its present levels and under normal operating conditions is well within the prescribed conservative limits for protection against external and internal beta radiation. There is also the tentative evidence discussed above that long-lived gamma activity retained within the body does not result from association with Naval nuclear power plants but rather from the general slight increase in the background effects from nuclear fallout. During certain periods of weapon testing, it seems likely that nuclear submarines, with their closely controlled radiation levels and the shielding effects of the surrounding sea, are exceptionally safe environments from the standpoint of exposure to ionizing radiation.

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BIBLIOGRAPHY

1. Miles, M. E., and Iltis, T. J., Radiological Controls For Naval Nuclear Propulsion Plants, (NAVSHIPS 389-0153), Bureau of Ships, Navy Department, December, 1959.

2. Christianson, C., and Maggio, R. C., Counting Techniques, Procedures and Instrumentation, (NAVSHIPS 93393A), Bureau of Ships, Navy Department, June, 1960.

3. National Bureau of Standards Handbook Number 51, Radiological Monitoring Methods and Instruments, United States Department of Commerce, 7 April 1952.

4. Cohn, S. H., Lippincott, S. W., Cronkite, E. P., and Reizenstein, P. G., Application of Whole-Body Gamma Spectrometry to Clinical Tracer Methods, Medical Research Center, Brookhaven National Laboratory, Upton, Long Island, New York. Unpublished paper.

5. Cohn, Stanton H., Ph.D., Medical Physics Division, Medical Department, Brookhaven National Laboratory, Upton, Long Island, New York. Personal communication of 7 September 1962.